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13. SUPPLEMENTARY NOTES The U. S. Government is joint author of this work and has the right to use, modify, reproduce, release, perform, display, or disclose the work. Published in © 2006 IEEE. Cleared for Public Release by ESC/PA number: ESC-06-0561. In a previous paper, we described a method for growing thick epitaxial layers of GaAs on orientation-patterned wafers by low pressure hydride vapor phase epitaxy. The low pressure method allows for rapid growth at rates well above 100 µm/hr and layers up to 1 mm thick have been successfully produced. In this paper we present characterization of these layers by optical microscopy, Hall measurement, and cathodoluminescence imaging. We demonstrate growth of low free carrier concentration, mm-thick orientationpatterned GaAs for efficient nonlinear optical conversion.					
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CHARACTERIZATION OF HVPE-GROWN THICK GaAs STRUCTURES FOR IR AND THz GENERATION

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Abstract

In a previous paper, we described a method for growing thick epitaxial layers of GaAs on orientation-patterned wafers by low pressure hydride vapor phase epitaxy. The low pressure method allows for rapid growth at rates well above 100 $\mu\text{m/hr}$ and layers up to 1 mm thick have been successfully produced. In this paper we present characterization of these layers by optical microscopy, Hall measurement, and cathodoluminescence imaging. We demonstrate growth of low free carrier concentration, mm-thick orientation-patterned GaAs for efficient nonlinear optical conversion.

I. Background

Hydride vapor phase epitaxy (HVPE) has been applied to the growth of thick quasi-phase-matched GaAs for nonlinear optical applications. Quasi-phase-matched (QPM) GaAs is being investigated as the active material for IR and THz generation using processes such as second harmonic generation, optical parametric oscillation and difference frequency generation. (1,2) Quasi-phase-matching is accomplished by varying the nonlinear optical susceptibility periodically across the crystal, using a method developed at Stanford University (3) in collaboration with AFRL. The GaAs crystal structure is polar, and therefore the nonlinear optical susceptibility can be modulated by periodically reversing the orientation of the GaAs crystal structure. Such modulation can be created epitaxially, using a non-polar Ge layer to reverse the GaAs orientation, and subsequent lithography and etching to periodically reveal the substrate, forming a grating of alternating crystallographic orientation

(Fig. 1). The grating must be grown very thick to accommodate the pump laser, which enters at the side of the grating and propagates across the structure.

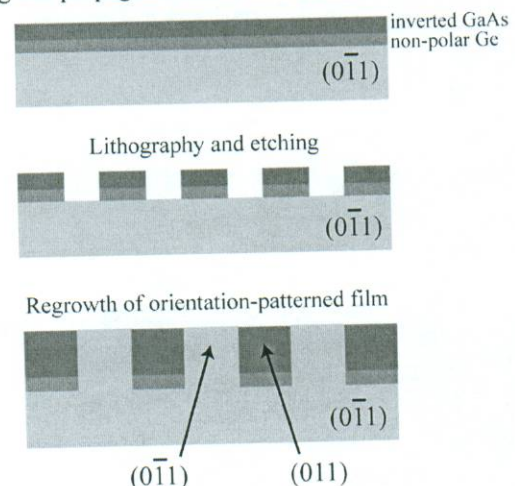


Fig. 1. Schematic of the fabrication of orientation-patterned templates for thick HVPE growth.

In this work we focus on growth-related issues influencing the efficiency of signal generation in orientation-

patterned GaAs (OP-GaAs). For high efficiency, we require low optical loss, low free carrier concentration, and propagation of the patterned grating through the thick (~1 mm) epitaxial growth. We have investigated these factors using optical and electrical characterization.

The requirement for mm-thick epitaxial layers drives the use of HVPE over other technologies such as molecular beam epitaxy (MBE) and organometallic vapor phase epitaxy (OMVPE). Although MBE is used for the initial growth of the patterned template, it is not well-suited for the production of mm-thick layers. We have demonstrated thick growth of homoepitaxial GaAs via HVPE in a custom-built reactor at the Air Force Hanscom Research Site. (4) The system consists of a horizontal quartz tube, heated by a three-zone furnace and sealed to allow low pressure operation in the range of 1 to 5 torr (Fig. 2). HCl vapor passing over a liquid Ga source reacts to form GaCl, which is transported to the substrate. Arsenic is supplied in the form of arsine (AsH_3), which decomposes on the surface. Both the Ga source and the substrate are heated to temperatures in the range of 650 to 750°C; the temperature gradient along the tube drives GaCl formation at the source and GaAs deposition on the substrate. The growth rate can be controlled by varying the vapor supersaturation – i.e., increasing or decreasing the partial pressures of the reactants with respect to their equilibrium values alters the tendency toward growth or etching of GaAs. Using HVPE we can attain growth rates of 150 microns per hour; however, careful control over the gas supersaturation and furnace temperature profile is necessary to maintain this growth rate for many hours, while preventing parasitic GaAs deposition on the reactor walls upstream of the sample. Using such controls (4), we achieve thicknesses of over 800 μm in a single 8 hour growth run and attain thicknesses over 1 mm total after multiple runs.

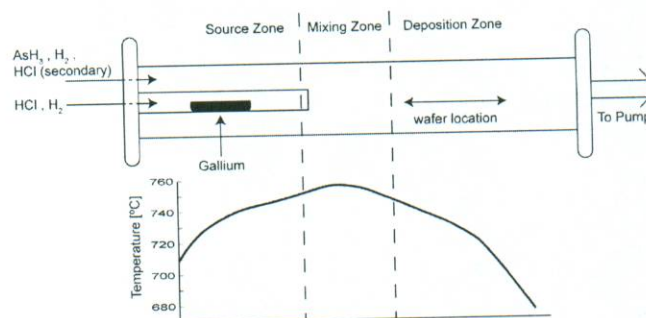


Fig. 2. Schematic of low-pressure hydride vapor phase epitaxial reactor.

II. Grating Structure

It is necessary that the patterned structure on the template is propagated cleanly (Fig. 3) for the full thickness of the layer since the grating periodicity provides the quasi-phase-matching. There is a tendency for the domains to overgrow one another, resulting in a partial or complete loss of the orientation-patterned structure. The overgrowth is significantly worse when the grating walls are perpendicular to the substrate tilt (usually 4° towards the (111)B); however, even when the grating walls are aligned with the substrate tilt we observe some overgrowth. An example of such a film is shown in Fig. 4 – a 1.1 mm thick GaAs grating resulting from a three-step growth with intermediate polishing. The domain walls begin to bend near the end of the growth, and with continued growth the (011) domains will disappear. We associate this overgrowth with the step-flow growth mechanism because the zone of overgrowth sweeps across the wafer in the direction of step-flow (i.e., opposite the tilt direction). The orientation that succeeds in such cases is always the orientation corresponding to that of the substrate, which exists in alternating domains and in the case of Fig 4, also exists in the unpatterned regions that surround the gratings.



Fig. 3 Cleaved and etched cross-section of HVPE growth on an OP-GaAs template. This image shows straight domain walls resulting from uniform propagation of the template pattern in the [100] growth direction.

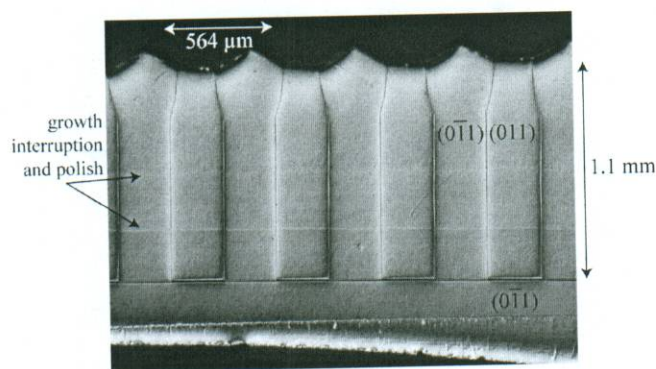


Fig. 4. 1.1 mm-thick GaAs HVPE layer grown in 3 steps on an orientation-patterned template.

III. Defect Characterization

Free carrier absorption is a dominant mechanism of optical absorption in the far-IR. We measure free carrier concentration and mobility by Hall effect using GaAs layers grown on semi-insulating substrates. Control over HVPE reactor temperatures and HCl flow rate has resulted in a decrease in free carrier concentration from $n \sim 10^{16} \text{ cm}^{-3}$ to $p \sim 10^{13} \text{ cm}^{-3}$ and allows control over the type and concentration of free carriers (Fig. 5). Mobility measurements suggest that these layers are compensated. Increasing the HCl supply is believed to reduce the unintentional incorporation of Si generated by the reaction between the quartz reactor tube and the HCl. (5) Our observations are consistent with this model.

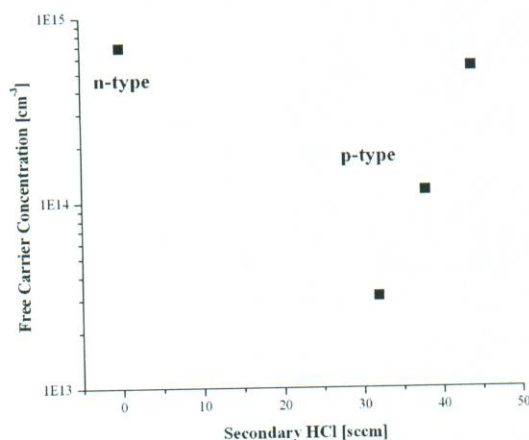


Fig. 5. Variation in free carrier type and concentration with flow rate of secondary HCl.

To better understand the defects controlling the electrical behavior and to investigate other defects which may lead to optical loss, HVPE-grown GaAs and OP-GaAs were characterized by spectral imaging cathodoluminescence (CL), using a Gatan XiCLone system attached to a JEOL 820 scanning electron microscope. Panchromatic and spectrum images were acquired at low temperatures (80K). In the CL spectra of the layer (Fig. 6(a)), the main band indicates a high quality film with few defects, with small compressive stress. Bands at 911 nm (1.36 eV) and 930 nm (1.33 eV) are associated with Si complexes ($\text{As}_{\text{Ga}}\text{-Si}_{\text{Ga}}$) and Ga vacancies, (V_{Ga}), respectively. Correlation with Hall measurements shows an inverse relationship between the relative intensity of these extrinsic bands and the free electron concentration. These bands are observed in both types of domains, with a higher relative intensity in the (011) oriented domains. Defect incorporation at different rates is likely a consequence of the faceted surface that evolves during growth. It has been established that impurity incorporation depends on crystallographic orientation during VPE (6), and therefore domains with (111) faceted surfaces will have different defect densities than domains with flat (001) surfaces.

The CL images reveal a number of features related to defects in the orientation-patterned structure (Fig. 6(b)). The walls separating the domains are revealed as dark lines, indicating non-radiative recombination, likely because the antiphase bonds (As-As and Ga-Ga) act as mid-gap levels. In some images cross-hatched lines are revealed in the domains. These lines show arsenic antisite defects (As_{Ga}) formed during dislocation glide on $\{111\}$ planes. Growth conditions which minimize the width of the domain wall region and the formation of dislocations should be investigated, due to the potential for optical loss.

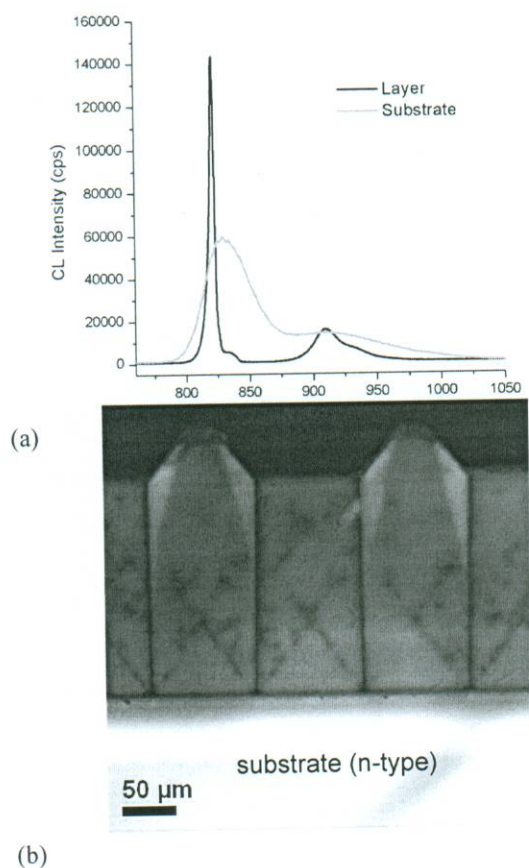


Fig. 6 CL spectra from OP-GaAs layer and substrate (a) and panchromatic 80K CL image of OP-GaAs grating (b).

IV. Conclusion

Hydride vapor phase epitaxy has been applied to the growth of mm-thick orientation-patterned GaAs layers for nonlinear optics. Conditions suitable for the growth of films of thicknesses over 1 mm have been established. The template pattern must be propagated through this thickness, to achieve the maximum conversion efficiency. Characterization by Hall measurement and cathodoluminescence has revealed the influence of processing on defects related to optical loss mechanisms.

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References

1. G. Imeshev, M. E. Fermann, K. L. Vodopyanov, M. M. Fejer, X. Yu, J. S. Harris, D. Bliss and C. Lynch, to be published in Optics Express.
2. P. S. Kuo, K. L. Vodopyanov, M. M. Fejer, D. M. Simanovskii, X. Yu, J. S. Harris, D. Bliss, and D. Weyburne, Optics Letters, **31**, 71 (2006).
3. C. B. Ebert, L. A. Eyres, M. M. Fejer and J. S. Harris, Jr., J. Crystal Growth **201/202**, 187 (1999); L. A. Eyres et al., Appl. Phys. Lett. **79**, 904 (2001).
4. D. F. Bliss, C. Lynch, D. Weyburne, K. O'Hearn, and J.S. Bailey, J. Crystal Growth, **287** 673 (2006).
5. J. V. DiLorenzo and G. E. Moore, Jr., J. Electrochem. Soc. **118**, 1823 (1971).
6. J. V. DiLorenzo, J. Crystal Growth **17**, 189 (1972).